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<b>(51) International Patent Classification <sup>5</sup> :</b> B32B 7/12 cog j 123/20, 123/16 123/04	A1	<b>(11) International Publication Number:</b> WO 93/17863 <b>(43) International Publication Date:</b> 16 September 1993 (16.09.93)
<b>(21) International Application Number:</b> PCT/US93/02016 <b>(22) International Filing Date:</b> 4 March 1993 (04.03.93)  <b>(30) Priority data:</b> 848,450 9 March 1992 (09.03.92) US  <b>(71) Applicant:</b> MOBIL OIL CORPORATION [US/US]; 3225 Gallows Road, Fairfax, VA 22037-0001 (US). <b>(72) Inventors:</b> O'BRIEN, Jeffrey, James ; 3215 Woods Edge, Walworth, NY 14568 (US). SEXTON, Donald, Francis ; 108 Blackwatch Trail, Fairport, NY 14450 (US). <b>(74) Agents:</b> ROBERTS, Peter, William et al.; Mobil Oil Corporation, 3325 Gallows Road, Fairfax, VA 22037-0001 (US).		<b>(81) Designated States:</b> AU, CA, JP, KR, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> HEAT SEALABLE THERMOPLASTIC FILMS  <b>(57) Abstract</b> <p>A base thermoplastic film has a coextruded or coated heat seal layer formed from a polymer blend which provides good peel-apart heat seals. When the film is sealed to itself, the seal opens by separation at the original seal surface without film tear, or by delamination of the sealant material from the primary HDPE film substrate without the formation of strands extending between the two base film. The heat seal layer which may be on one or both sides of the base layer comprises a ternary blend of a random ethylene-propylene copolymer, a butene polymer and a low density polyethylene. The films are cast, and oriented in the solid state up to about two times in the machine direction and six times or more in the transverse direction to give films having good dead-fold and water vapor transmission rate characteristics making them highly suited for packaging, particularly for dry foods.</p>		

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low density, medium density, linear low density polyethylenes and mixtures thereof may also be used.

According to the present invention, the base film is provided with a heat seal layer which comprises a blend of three components which are formulated to ensure that each component is present in an amount which effectively interferes with the seal strength development of any other single component: a controlled seal strength is generated by the interaction of the three components, resulting in a heat seal which is strong enough to provide an adequate seal which can, however, be peeled apart without tearing the base film. Seal strengths of 100 to 500 grams per inch which permit ready peel-apart separation can be obtained with seal temperatures in the range of about 170° to about 250°F (measured with a WrapAide Crimp Sealer run at 20 psi, 0.75 second dwell time). In commercial operation, sealing may be successfully achieved over a wide range of temperatures suitable for commercial equipment while retaining the desired peel separation characteristics.

The sealant blends are composed of at least one component from each of the three groups (A, B, C) below.

The Group A polymeric component is a random ethylene-propylene copolymer. This may be derived from ethylene and one or more co-monomers. The propylene content of these random copolymers is typically from 70 to 85 weight percent, more usually from 75 to 85 percent, with the balance of ethylene and any other comonomers such as butylene. Suitable copolymers of this type are random copolymers of ethylene and propylene or random terpolymers of ethylene, propylene and butylene. Preferred copolymers of this type include the following:

Ethylene-propylene copolymers containing 2 - 10 weight percent random ethylene, e.g. 3 - 7 weight percent ethylene.

5 Ethylene-propylene-butylene random terpolymers containing 1 - 5 weight percent random ethylene, 10 - 25 weight percent random butylene. The amounts of the random ethylene and butylene components in these copolymers are typically in the range of 10 to 25 percent total (ethylene plus butylene). Typical  
10 terpolymers of this type include those with about 1 - 5 percent ethylene and 10 - 25 percent butylene.

These copolymers typically have a melt flow rate in the range of about 5 to 10 with a density of about 0.9 and a melting point in the range of about 115° to  
15 about 130°C.

The Group B polymer component comprises a low density polyethylene. This may be a linear low density polyethylene (LLDPE) or a non-linear polyethylene. These polymers typically have a melt  
20 index of 1 to 5. The low density polyethylenes should have a density of 0.88 to 0.93 while the linear materials may have a density as high as 0.94, usually in the range 0.90 - 0.94, e.g. 0.918 or 0.921, with a melt index from 1 to 5. The linear low density  
25 polyethylenes may be derived from ethylene together with other higher comonomers such as hexene-1 or octene-1.

The Group C polymer component is a butene polymer which may be a homopolymer or copolymer with minor  
30 amounts of comonomers such as ethylene and/or propylene. These butene polymers typically have a molecular weight of at least 10,000 and a melt index from 1 to 6, usually from 2 to 4.

The three components of the composition are used  
35 in a blend which is formulated to provide a seal

strength of 100 to 500 grams/inch (measured with a WrapAide at a seal temperature of 170° to 250°F, at 20 psi and a dwell time of 0.75 seconds). The best results are obtained when the three components are in approximate balance with one another, i.e. with no more than 50 weight percent of the total blend from one component. This implies that each component will be present in an amount from 10 to 50 weight percent, usually 15 to 45, typically 20 to 40, weight percent of the total blend. As shown below, blends which contain about 20 weight percent of one of the three components with the balance made up of equal amounts of the other two components, give very favorable results.

Thus it is desirable that the three components comprise a blend of 10 to 50% of a random copolymer of ethylene and propylene, from 10 to 50% of low density polyethylene and from 10 to 50% of polybutene; or a blend of 10 to 50% of a random terpolymer of ethylene, propylene and butylene, from 10 to 50% of low density polyethylene and from 10 to 50% of polybutene; or a blend of 30 to 50% of the random copolymer of ethylene and propylene, from 10 to 30% of low density polyethylene and from 10 to 30% of polybutene; or a blend of 10 to 30% of the random copolymer of ethylene and propylene, from 10 to 30% of low density polyethylene and from 30 to 50% of polybutene; or a blend of 30 to 50% of the random terpolymer of ethylene, propylene and butylene, from 10 to 30% of low density polyethylene and from 10 to 30% of polybutene; or a blend of 10 to 30% of the random terpolymer of ethylene, propylene and butylene, from 30 to 50% of low density polyethylene and from 10 to 30% of polybutene; or a blend of 10 to 30% of the random terpolymer of ethylene, propylene and butylene,

from 10 to 30% of low density polyethylene and from 30 to 50% of polybutene.

5 The polymer components of all three groups may contain stabilizers to minimize oxidative and thermal degradation, as well as other additives to achieve other functionality including static reduction, ease of processing, and ink receptivity. Silicone oil may be added, e.g. in amounts from 0.2 to 2 weight percent to improve slip characteristics and ease of processing; polydimethylsiloxane is preferred for this purpose. Wax, preferably Fischer-Tropsch or microcrystalline wax, may be added to the blends in an amount typically up to about 10 weight percent for improved WVTR performance.

15 The heat seal compositions are used on HDPE films which have been biaxially oriented to provide good WVTR properties as well as other characteristics which are desirable in packaging films. The seal blends may be used on one or both sides of the base HDPE films.

20 Where the base film is HDPE, the base film is preferably biaxially oriented to a deformation ratio from 1.1:1 to 2:1, usually from 1.25:1 to 2:1, in the machine direction (MD), and to a deformation ratio from 6:1 to 12:1 in the transverse direction (TD).  
25 These films, preferably not having a thickness of more than 2.5 mils, have reduced water vapor transmission (WVTR), improved dead-fold, and other physical properties which are markedly better than blown HDPE films, even when the total film thickness is reduced to less than 1 or 2 mils. When provided with a  
30 blended heat-seal layer of the ethylene-propylene random copolymers the films are particularly suited for use in packaging, especially of dry foodstuffs. The films may be used in a wide variety of packaging equipment including vertical form, fill and seal  
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(VFFS) and high speed horizontal wrapping machines.

These biaxially oriented films are made using a major proportion of a high density polyethylene (HDPE) having a density of at least 0.96. The film can be composed exclusively of a single HDPE resin, a mixture of HDPE resins, or of HDPE containing a minor proportion of other resource polymers. These high density polyethylenes typically have a melt index from 0.5 to 10, usually from 0.7 to 2. The mixture of HDPE resins gives better processing characteristics in the extruder by reducing extruder torque. Films made with a blend of HDPE resins reduce splittiness of the film which manifests itself as the tendency of the film to break in the TD direction during operation on vertical, form, fill and seal (VFFS) machinery.

The blends of HDPE polymers can comprise two or more polymers all of which preferably have densities of at least 0.96. Blends of HDPE polymers advantageously comprise a major proportion of HDPE having a melt index of 0.5 to 6 and one or more polymers having a different melt index.

Terblends have been found particularly desirable. Suitable terblends generally comprise 50 to 98 weight percent, preferably 84 to 96 weight percent of HDPE having a density of at least 0.96 and a melt index of greater than 0.5 to 2; 1 to 25 weight percent, preferably 3 to 8 weight percent of HDPE having a density of at least 0.96 and a melt index of 0.1 to 0.5; and 1 to 25 weight percent, preferably 3 to 8 weight percent, of HDPE having a density of at least 0.96 and a melt index of greater than 2 to 8. Preferably, the second and third HDPE polymers which are minor components are present in about equal amounts.

The heat seal layer can be applied to the HDPE base film in any conventional manner, for example, by coating or coextrusion before orientation or by coating the HDPE after one or both of the biaxial orientation operations.

The base films of this invention can include other polymer layers in addition to the heat seal layer, for example, polymers having barrier properties for gases such as oxygen.

The proper degree of orientation in the film provides the desired physical properties, as well as good WVTR and dead-fold characteristics. For example, it has been determined that films with a thickness of 1.4 to 4 mils will have acceptable WVTR ( $\text{g-mil}/100 \text{ in}^2/24 \text{ hr} - 1 \text{ atm}$ ) of less than about 0.2/mil whereas a somewhat heavier gauge (at least 1.5 times thicker) is needed in a blown HDPE film to achieve the same WVTR. The benefits of reduced WVTR are due to the improvements obtained by biaxial orientation below the HDPE melting point. Although higher density HDPE resin having a density of at least 0.957 can be made directly into thin films by cast extrusion, problems of curling, uniformity, flatness and high WVTR remain as obstacles. Accordingly, thin HDPE films of 0.8 to 1.5 mils having the best balance of properties, particularly for VFFS applications, are obtained with imbalanced biaxially oriented films prepared from films having a cast gauge of 12 to 20 mils reduced to the desired gauge by orientation. The final film gauge will typically be not more than about 2.5 mils.

The films may be produced and oriented in the conventional manner. When the heat sealable layer is present on one or both sides of the HDPE film, cast extrusion is generally accomplished using a multi-roll stand system having three or more rolls.

In the usual manner the film is heated to its orientation temperature and first subjected to MD orientation between two sets of nip rolls the second rotating at a greater speed than the first in an amount equal to the desired draw ratio. Then the film is TD oriented by heating and subjecting it to transverse stretching in a tenter frame. Typically MD orientation is conducted at 60° to 120°C and TD orientation at 110° to 145°C.

The invention is illustrated by the following Examples in which all parts are by weight unless otherwise specified.

#### EXAMPLES 1-3

These examples used a heat seal blend comprised of:

a random ethylene, propylene, butene-1 terpolymer with a weight ratio of 1.5 - 3% ethylene and 12 - 18% butene-1, a low density polyethylene (LDPE) with a density of .92 and a melt index of 2.2, and a polybutene-1 (PB) with a melt index of 4.

The amounts of the blend components were varied as shown below:

<u>Blend Component</u>	<u>Terpolymer</u>	<u>LDPE</u>	<u>PB</u>
Example 1	20	40	40
Example 2	40	20	40
Example 3	40	40	20

Two layer biaxially oriented films having final thicknesses in the range of 0.4 to 5 mils were prepared by coextruding a primary HDPE layer comprising 90% of the film thickness with the sealant layer. The primary layer HDPE had a broad molecular weight distribution and a density of 0.96 with a melt index of about 1.

The films were prepared in a three roll stack system. The films were then oriented 1.3 times in the MD at about 115°C, and 10 times in the TD direction 115°-160°C in a tenter frame.

The films were run on a horizontal wrapping machine producing slugs of wrapped crackers. They performed well in crimp sealing at the temperatures shown below, providing seal strengths of 100-500 grams per inch, with seals peeling open without tearing the film.

	<u>Seal Temperature, °F</u>	<u>WrapAide*, °F</u>
Example 1	200-300	170-240
Example 2	200-290	170-230
Example 3	200-280	180-230

\*20 psi, 0.75 sec. dwell time

The WVTR in all cases was 0.18-0.22 g-mil/100 in<sup>2</sup>/24 hr at 1 atmosphere.

#### EXAMPLES 4-6

These examples used a heat seal blend comprised of:

a random ethylene-propylene copolymer containing 4 - 8% random ethylene, a low density polyethylene (LDPE) with a density of .92 and a melt index of 2.2, and a polybutene-1 (PB) with a melt index of 2.

The amounts of the blend components were varied as shown below:

<u>Blend Component</u>	<u>Copolymer</u>	<u>LDPE</u>	<u>PB</u>
Example 4	20	40	40
Example 5	40	20	40
Example 6	40	40	20

A two layer biaxially oriented film having a final thickness of 1.15 mils was prepared by coextruding a primary HDPE layer comprising 90% of the film thickness with the sealant layer. The primary

layer HDPE had a broad molecular weight distribution and a density of 0.96 with a melt index of 1.

The films were prepared in a three roll stack system. The films were then oriented 1.3 times in the MD at about 115°C, and 10 times in the TD direction 115°-160°C in a tenter frame.

The films were run on a horizontal wrapping machine producing slugs of wrapped crackers. They performed well in crimp sealing at the temperatures shown below, providing seal strengths of 100-500 grams per inch, with seals peeling open without tearing the film.

	<u>Seal Temperature, °F</u>	<u>WrapAide, °F*</u>
Example 4	220-280	180-230
Example 5	220-270	180-200
Example 6	220-300	180-230

\* 20 psi, 0.75 sec. dwell time

The WVTR in all cases was 0.18-0.22 g-mil/100 in<sup>2</sup>/24 hr at 1 atmosphere.

#### EXAMPLES 7-9

These examples used a heat seal blend comprised of:

a random ethylene, propylene, butene-1 terpolymer with a weight ratio of 1 - 2% ethylene and 20 - 25% butene-1, a linear low density polyethylene (LLDPE) with a density of .92 and a melt index of 2.2, and a polybutene-1 (PB) with a melt index of 4.

The amounts of the blend components were varied as shown below:

<u>Blend Component</u>	<u>Copolymer</u>	<u>LLDPE</u>	<u>PB</u>
Example 7	20	40	40
Example 8	40	20	40
Example 9	40	40	20

Two layer biaxially oriented films having final thicknesses in the range of 0.4-5 mils were prepared

by coextruding a primary HDPE layer comprising 90% of the film thickness with the sealant layer. The primary layer HDPE had a broad molecular weight distribution and a density of 0.96 with a melt index of 2.

The films were prepared in a three roll stack system. The films were then oriented 1.3 times in the MD at about 115°C, and 10 times in the TD direction 115°-160°C in a tenter frame.

The films were run on a horizontal wrapping machine producing slugs of wrapped crackers. They performed well in crimp sealing at the temperatures shown below, providing seal strengths of 100-500 grams per inch, with seals peeling open without tearing the film.

	<u>Seal Temperature, °F</u>	<u>WrapAide*, °F</u>
Example 7	230-280	180-240
Example 8	200-270	180-220
Example 9	230-300	190-240

\* WrapAide 20 psi, 0.75 sec. dwell time

The WVTR in all cases was 0.18-0.22 g-mil/100 in<sup>2</sup>/24 hr at 1 atmosphere.

#### EXAMPLES 10-11

These examples used a heat seal blend comprised of:

a random ethylene, propylene, butene-1 terpolymer with a weight ratio of 1.5 - 3% ethylene and 12 - 18% butene-1, a low density polyethylene (LDPE) with a density of .92 and a melt index of 2.2, and a polybutene-1 (PB) with a melt index of 4.

The amounts of the blend components were varied as shown below:

<u>Blend Component</u>	<u>Copolymer</u>	<u>LLDPE</u>	<u>PB</u>
Example 10	60	20	20
Example 11	40	0	40

Two layer biaxially oriented films having final thicknesses of in the range of 0.4-5 mils were prepared by coextruding a primary HDPE layer comprising 90% of the film thickness with the sealant layer. The primary layer HDPE had a broad molecular weight distribution and a density of 0.96 with a melt index of 1.

The films were prepared in a three roll stack system. The films were then oriented 1.3 times in the MD at about 115°C, and 10 times in the TD direction 115°-160°C in a tenter frame.

The WVTR in all cases was 0.18-0.22 g-mil/100 in<sup>2</sup>/24 hr at 1 atmosphere.

The films were run on a horizontal wrapping machine producing slugs of wrapped crackers. The films of Example 10 performed well in crimp sealing temperatures of 200-210°F (170-190 on the WrapAide at 20 psi, 0.75 sec. dwell time), providing seal strengths of 100 - 400 g./in but the temperature range is too narrow for commercial operation (a minimum 30°F range, and preferably 40°F, is desired for commercial operation). At higher seal temperatures above 220°F, seal strength increased to values of 400 - 1,000 grams per inch, and the base film tore when the packages were opened.

The films of Example 11 performed well at crimp sealing temperatures of 200-210°F (170-190 on the WrapAide at 20 psi, 0.75 sec. dwell time), providing seal strengths of 100 - 400 g./in but the temperature range is too narrow for commercial operation. At higher seal temperatures, seal strength increased to values of 400 - 900 grams per inch, and the base film tore when the packages were opened.

CLAIMS:

1. A film structure comprising a thermoplastic base layer having a heat sealable layer on at least one side which, on heat sealing, forms a seal which is separable at the original seal surface without film tear, or by delamination of the sealant material from the base layer.
2. A film structure according to claim 1 in which the heat sealable layer has a seal strength of 100 to 500 grams per inch (170° to 250°F seal temperature, 20 psi, 0.75 sec. dwell time).
3. A film structure according to claim 1 or 2 in which the heat sealable layer comprises a ternary blend of:
  - a) a random copolymer of ethylene and propylene and/or a random terpolymer of ethylene, propylene and butylene; and
  - b) low density polyethylene; and
  - c) polybutene,there being no less than 10 wt%, and no more than 50 wt%, of any one component a), b) or c) present in the blend, the weight percentage being by weight of the total blend.
4. A film structure according to claim 3 in which one of the components a), b) or c) is present in the blend in an amount from 30 to 50 wt%, the other two components each being present in the blend in an amount from 10 to 30 wt%, the weight percentage being by weight of the total blend.

5. A film structure according to claim 3 or 4 in which the random copolymer of ethylene and propylene has a propylene content from 75 to 85 weight percent.
- 5 6. A film structure according to any one of claims 3, 4 or 5 in which the random copolymer of ethylene and propylene has a random ethylene content from 2 to 10 weight percent.
- 10 7. A film structure according to claim 3 or 4 in which random terpolymer of ethylene, propylene and butylene has a random ethylene content from 1 to 5 weight percent and a random butylene content from 10 to 25 weight percent.
- 15 8. A film structure according to any one of claims 3 to 7 in which component a) has a melt flow rate from 5 to 10 with a density of 0.9 and a melting point from 115° to 135°C.
- 20 9. A film structure according to any one of claims 3 to 8 in which the low density polyethylene component comprises a linear low density polyethylene.
10. A film structure according to any preceding claim wherein the base layer comprises high density polyethylene (HDPE).
- 25 11. A film structure according to claim 10 wherein the HDPE has a density of at least 0.96.

12. A film structure according to claim 10 in which the base layer comprises an HDPE blend of two or more polymers each of which has a density of at least 0.96.

5 13. A film structure according to claim 12 in which the HDPE blend comprises a major proportion of HDPE having a melt index of 0.5 to 10.

14. A film structure according to claim 12 or 13 in which the HDPE blend comprises a terblend of:

10 i) 50 to 98 weight percent of HDPE having a density of at least 0.96 and a melt index greater than 0.5 to about 2;

15 ii) 1 to 25 weight percent of HDPE having a density of at least 0.96 and a melt index from 0.1 to 0.5;

iii) 1 to 25 weight percent of HDPE having a density of at least 0.96 and a melt index greater than 2 to 8, the weight percentage being by weight of the total HDPE blend.

20 15. A film structure according to any one of claims 10 to 14 wherein the base layer is oriented in the solid state to a deformation ratio from 1.1:1 to 2:1 in the machine direction, and to a deformation ratio from 6:1 to 12:1 in the  
25 transverse direction.

16. A heat seal composition for oriented films which comprises a blend of components a), b) and c) as defined in claim 3 or 4.

30 17. Use of a composition defined in claim 16 for forming heat seals in biaxially oriented film structures comprising high density polyethylene.

**Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)**

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.: 17  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:  
  
Claim 17 is directed to a "use" PCT Article 17(2)(a)(i). "Use" claims not in method terminology are not searched by this authority.
3. ☒ Claims Nos.: 5-17  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

**Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)**

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐

The additional search fees were accompanied by the applicant's protest.

☐

No protest accompanied the payment of additional search fees.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US93/02016

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(5) :B32B 7/12

US CL :428/349, 516; 525/240

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/349, 516; 525/240

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US, A, 4,921,749 (BOSSAERT ET AL) 01 MAY 1990, see Abstract.	1, 2
X	US, A, 4,275,120 (WEINER) 23 JUNE 1981, entire document.	1-4



Further documents are listed in the continuation of Box C.



See patent family annex.

* "A"	Special categories of cited documents: document defining the general state of the art which is not considered to be part of particular relevance	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"E"	earlier document published on or after the international filing date	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"O"	document referring to an oral disclosure, use, exhibition or other means	"Z"	document member of the same patent family
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search

06 MAY 1993

Date of mailing of the international search report

24 JUN 1993

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